Viscosity of argon and of argon-ammonia mixtures under pressures

AUTHOR(S):

Hongo, Masaru

CITATION:

Hongo, Masaru. Viscosity of argon and of argon-ammonia mixtures under pressures. The Review of Physical Chemistry of Japan 1979, 48(2): 63-71

ISSUE DATE:

1979-02-10

URL:

http://hdl.handle.net/2433/47064

VISCOSITY OF ARGON AND OF ARGON-AMMONIA MIXTURES UNDER PRESSURES*

BY MASARU HONGO

The viscosity of argon and of four argon-ammonia gaseous mixtures was measured using an oscillating disk viscometer at temperatures, 25, 50, 75, and 100°C; for argon, at the pressure range up to about 120×10^5 Pa; and for argon-ammonia mixtures, up to around the saturated vapor pressure of ammonia at each temperature. The accuracy of the measurements was estimated to be within $\pm0.3\%$.

The gas viscosity values of dilute binary mixtures were calculated by applying a correction factor, which was set up by use of critical compressibility factors. The calculated values were in better agreement with the experimental ones than those calculated by original Chapman-Enskog's method.

The initial density dependence of the viscosity, $\eta_0^{-1}(\partial\eta/\partial\rho)_{\rho\to0}$, of argon-ammonia mixtures was obtained from the experimental values, and the effects of the concentration of nonpolar gases and of temperature on the initial density dependence were compared with those in the case of the nitrogen-ammonia system and of the hydrogen-ammonia system, where η is viscosity of gas, ρ density, and η_0 the viscosity extrapolated to zero density. It was concluded from the comparison that molecular association of polar gases was hindered by the presence of nonpolar molecules and by a rise in temperature, and that the larger the molecular weight of a nonpolar gas was, the more strongly the nonpolar molecules hindered moecular association of polar substances.

By taking into account the mole fraction of nonpolar gases, the initial density dependence of the viscosity could be correlated with the temperature reduced by the critical temperature of the mixtures, and a similar tendency was observed regardless of the system.

Introduction

This study is one of the researches to measure the gas viscosity of nonpolar-polar mixtures containing ammonia as a polar gas under pressures. The viscosities of the nitrogen-ammonia system[1] and of the hydrogen-ammonia system[2] were already determined, and the viscosities of argon and of argon-ammonia mixtures are presented in this paper. In order to investigate the effect of the molecular weight of a nonpolar gas on the viscosity of a polar gas, argon was used because its molecular weight is larger than that of nitrogen. The viscosity of the argon-ammonia system under pressures was measured by Iwasaki *et al.*[3] using an oscillating disk viscometer, but only at 20 and 30°C. The viscosity at atmospheric psessure was measured by Chakraborti *et al.*[4] (25 to 80°C)using a capillary-

(*Received September 30, 1978*)

* Presented in part at The 18th High Pressure Conference, Kyoto, Nov. 25th. 1977

1) M. Hongo and H. Iwasaki, *This Journal*, **47**, 90 (1977)

2) M. Hongo and H. Iwasaki, *ibid.*, **48**, 1 (1978)

3) H. Iwasaki, J. Kestin, and A. Nagashima, *J. Chem. Phys.*, **40**, 2988 (1964)

4) P.K. Chakraborti and P. Gray, *Trans. Faraday Soc.*, **61**, 2422 (1965)

flow viscometer and by Rakshit *et al.*[5] (−35 to 35°C) by the use of an oscillating disk viscometer.

In this study, the measurements of the viscosity of argon-ammonia mixtures were performed on a precision oscillating disk viscometer, which was the same as that previously used[1,2], at temperatures of 25, 50, 75, and 100°C, and over the range of pressure up to about 120×10^5 Pa for argon and up to around the saturated vapor pressure of ammonia at each temperature for argon-ammonia mixtures. The mole fractions of argon in the mixtures were approximately 0.2, 0.4, 0.6, and 0.8.

Experimental Method and Calculation of Viscosity

The gas viscosity was evaluated by the same method as that used by Iwasaki *et al.*[6] The empirical value C_N' of the apparatus constant, C_N, required for the evaluation of the viscosity was determined by the calibration measurements which were made using nitrogen at temperature of 25°C and at pressures below 25×10^5 Pa. The value of C_N' obtained from the calibration measurements was 1.1355 from 25 to 75°C, and 1.1378 at 100°C.

The density of argon was obtained through the *PV*-values by Michels *et al.*[7] and that of argon-ammonia mixtures was evaluated from the *Vri*-chart[8] in the same way as that used in the previous papers[1,2], where *Vri* is the ideal reduced molar volume.

Argon was supplied from the Nippon Oxygen Co., Ltd. and its purity was above 99.994%. Ammonia was purified through distillation as described before[1].

The experimental accuracy was estimated to be within ±0.3%.

Results

The viscosity values obtained for argon are shown in Table 1 and plotted as a viscosity *vs.* density diagram in Fig. 1. The deviations of the measured viscosity values from the smoothed curves are all within ±0.2%. The slopes of the viscosity isotherm, $(\partial\eta/\partial\rho)_T$, are always positive and almost the same in the experimental conditions, where η is viscosity of gas and ρ is density. The viscosity of argon under pressures, so far, was measured by Michels *et al.*[9] from 0 to 75°C below 2000×10^5 Pa; by Iwasaki *et al.*[10] from 25 to 150°C up to 130×10^5 Pa; by Flynn *et al.*[11] from −78 to 100°C below 200×10^5 Pa; by Reynes *et al.*[12] from 100 to 200°C up to 830×10^5 Pa; and by Gracki *et al.*[13] from −100 to 25°C below 170×10^5 Pa. These measurements were made using a capillary-flow viscometer, except

5) A.B. Rakshit, C.S. Roy, and A.K. Barua, *J. Chem. Phys.*, **59**, 3633 (1973)
6) H. Iwasaki and J. Kestin, *Physica*, **29**, 1345 (1963)
7) A. Michels, Hub. Wijker and Hk. Wijker, *ibid.*, **15**, 627 (1949)
8) K. Satoh, "Bussei Josu Suisanho", Maruzen, Japan (1954)
9) A. Michels, A. Botzen and W. Schuurman, *Physica*, **20**, 1141 (1954)
10) H. Iwasaki and H. Takahashi, *Bull. Chem. Res. Inst. Non-aqueous Solutions, Tohoku Univ.*, **6**,61 (1956)
11) G.P. Flynn, R.V. Hanks, N.A. Lemaire, and J, Ross, *J. Chem. Phys.*, **38**, 154 (1963)
12) E.G. Reynes and G. Thodos, *Physica*, **30**, 1529 (1964)
13) J.A. Gracki, G.P.Flynn and J. Ross, *J. Chem. Phys.*, **51**, 3856 (1969)

Table 1 Viscosity of argon

Pressure 10^5 Pa	Density 10^3 kg·m^{-3}	Viscosity 10^{-7} Pa·s	Pressure 10^5 Pa	Density 10^3 kg·m^{-3}	Viscosity 10^{-7} Pa·s
	25°C			75°C	
1.23	0.00197	226.1	1.26	0.00173	256.5
1.47	0.00236	226.6	1.26	0.00173	256.6
2.52	0.00405	226.7	2.03	0.00278	256.4
5.06	0.00812	226.8	5.17	0.00710	257.0
5.12	0.00822	226.9	10.11	0.01388	257.5
10.15	0.01632	228.1	20.37	0.02815	259.7
10.19	0.01637	227.9	41.38	0.05350	264.1
20.39	0.03323	229.9	61.11	0.08525	268.5
23.84	0.03919	231.0	82.43	0.1152	274.5
41.17	0.06797	235.4	102.81	0.14378	280.7
61.73	0.1030	242.0	113.39	0.16667	286.4
83.18	0.1398	249.7			
104.59	0.17696	258.6		100°C	
125.30	0.21291	267.9			
125.35	0.21296	268.2	1.23	0.00159	272.8
			1.36	0.00176	273.0
	50°C		2.03	0.00262	273.5
			5.06	0.00654	273.6
1.24	0.00183	242.0	10.11	0.01308	274.5
1.52	0.00225	242.1	20.27	0.02614	276.4
2.54	0.00228	242.0	40.44	0.05227	280.0
5.12	0.00757	242.8	61.05	0.07893	285.1
5.12	0.00757	242.7	82.12	0.1062	290.1
10.12	0.01498	243.5	102.60	0.13244	295.9
10.19	0.01508	243.5	122.01	0.15716	301.7
20.42	0.03054	245.4	125.98	0.16219	302.8
41.16	0.06212	250.0			
61.29	0.09299	255.6			
81.38	0.1240	261.8			
102.24	0.15638	270.1			
121.54	0.18535	276.8			
121.54	0.18595	277.5			

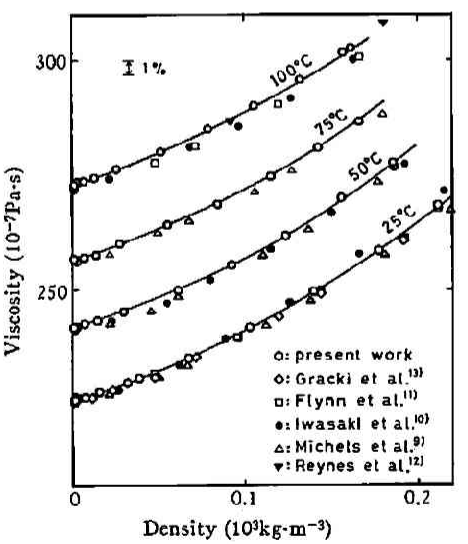

Fig. 1 Viscosity of argon

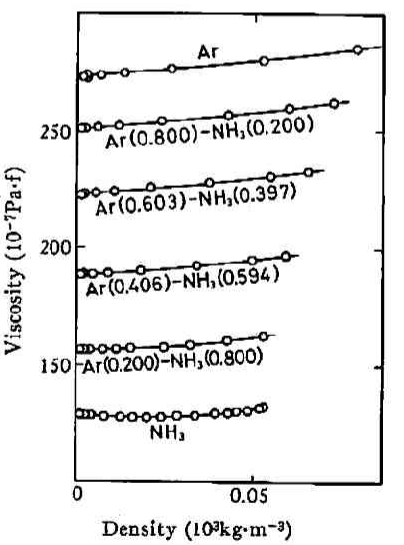

Fig. 2 Viscosity of argon-ammonia system at 100°C

that of Iwasaki *et al.*[10] who used an oscillating disk viscometer. As shown in Fig. 1, the viscosity values obtained in this work are in agreement with those obtained in the investigations mentioned above within the experimental error.

The viscosity measurements for four argon-ammonia mixtures were carried out at the same temperatures as for argon and at pressures up to around the saturated vapor pressure of ammonia at each temperature, *i.e.*, about 10×10^5 Pa at 25°C, 20×10^5 Pa at 50°C, 35×10^5 Pa at 75°C, and 60×10^5 Pa at 100°C. The viscosity values obtained for the mixtures are given in Table 2. The results obtained for the gaseous mixtures at 100°C are shown in Fig. 2. The viscosity values of pure ammonia shown in Fig. 2 are those reported in Ref. 1. As shown in Fig. 2, the initial slope of the viscosity-density curves for the gaseous mixtures, $(\partial\eta/\partial\rho)_{\rho\to0}$, increases as the mole fraction of argon increases, and becomes nearly constant above 0.406 mole fraction of argon.

Table 2 Viscosity of argon-ammonia mixtures

Pressure 10^5 Pa	Density 10^3kg·m^{-3}	Viscosity 10^{-7}Pa·s
25°C		
Ar(0.208)-NH$_3$(0.792)		
1.23	0.00108	129.5
1.33	0.00117	129.5
2.53	0.00223	129.5
4.54	0.00399	129.2
6.65	0.00591	129.0
8.67	0.00787	128.9
10.18	0.009425	128.8
Ar(0.397)-NH$_3$(0.603)		
1.23	0.00129	157.5
1.41	0.00149	157.7
3.06	0.00323	157.8
5.12	0.00539	158.0
7.10	0.00763	158.1
9.17	0.0100	158.1
10.68	0.01183	158.4
Ar(0.602)-NH$_3$(0.398)		
1.18	0.00146	184.8
1.28	0.00159	185.0
3.01	0.00374	185.1
5.16	0.00641	185.7
7.63	0.00961	186.0
9.90	0.0126	186.2
11.42	0.01467	186.4
Ar(0.800)-NH$_3$(0.200)		
1.25	0.00178	208.4
1.45	0.00207	208.2
2.99	0.00426	208.5
5.14	0.00733	208.9
7.15	0.0103	209.2
10.22	0.01489	209.9
12.35	0.01835	210.3
50°C		
Ar(0.211)-NH$_3$(0.789)		
1.17	0.000949	140.2
1.26	0.00102	140.4
2.03	0.00165	140.2
5.08	0.00420	140.2
10.17	0.008782	140.2
15.18	0.01349	140.2
20.39	0.01868	140.4
Ar(0.403)-NH$_3$(0.597)		
1.23	0.00120	168.3
1.42	0.00139	168.3
2.51	0.00246	168.4
5.09	0.00499	168.7
10.17	0.01028	169.2
15.14	0.01566	169.5
20.29	0.02154	170.0
21.21	0.02268	170.3
50°C		
Ar(0.600)-NH$_3$(0.400)		
1.24	0.00142	196.4
1.30	0.00149	196.7
2.02	0.00231	196.5
3.04	0.00349	196.8
5.12	0.00590	197.2
10.13	0.01179	197.8
15.26	0.01809	198.5
20.25	0.02440	199.6
22.69	0.02758	199.9
Ar(0.801)-NH$_3$(0.199)		
1.23	0.00161	220.9
1.26	0.00165	221.0
2.03	0.00267	221.1
3.05	0.00400	221.2
5.08	0.00668	221.5
10.13	0.01342	222.7
15.18	0.02034	223.3
20.32	0.02749	224.2
24.15	0.03285	225.0
75°C		
Ar(0.202)-NH$_3$(0.798)		
1.20	0.000895	148.0
1.25	0.000933	148.0
2.03	0.00152	148.0
5.06	0.00378	148.0
10.14	0.007830	148.1
20.28	0.01650	148.4
30.82	0.02642	149.1
35.76	0.03112	149.5
Ar(0.394)-NH$_3$(0.606)		
1.18	0.00106	177.6
1.24	0.00111	177.4
2.01	0.00181	177.7
5.08	0.00457	177.9
10.14	0.009363	178.3
20.34	0.01947	179.2
31.15	0.03080	180.6
36.74	0.03682	181.5
Ar(0.597)-NH$_3$(0.403)		
1.22	0.00129	208.2
1.37	0.00145	208.3
2.03	0.00215	208.6
5.07	0.00538	209.2
10.13	0.01089	209.8
20.27	0.02228	211.4
30.92	0.03482	213.2
37.43	0.04278	214.3
75°C		
Ar(0.794)-NH$_3$(0.206)		
1.24	0.00150	234.7
2.03	0.00247	234.7
5.09	0.00619	235.2
10.11	0.01231	236.1
15.23	0.01864	237.0
25.30	0.03136	238.9
36.16	0.04540	240.8
39.41	0.04958	241.5
100°C		
Ar(0.200)-NH$_3$(0.800)		
1.18	0.000819	156.3
1.26	0.000876	156.4
1.44	0.00100	156.5
2.03	0.00141	156.4
3.00	0.00209	156.5
5.07	0.00353	156.4
10.13	0.007260	156.5
15.15	0.01112	156.7
20.27	0.01512	157.0
31.76	0.02441	157.6
40.43	0.03193	158.7
51.06	0.04228	160.5
60.56	0.05269	162.5
Ar(0.406)-NH$_3$(0.594)		
1.24	0.00105	188.2
2.04	0.00173	188.3
5.05	0.00428	188.6
10.13	0.008730	189.0
20.29	0.01788	190.1
36.62	0.03362	192.2
51.70	0.04938	194.9
60.80	0.05907	196.8
Ar(0.603)-NH$_3$(0.397)		
1.22	0.00121	222.0
2.03	0.00202	222.2
5.05	0.00502	223.0
10.14	0.01019	223.5
20.29	0.02064	225.2
36.11	0.03771	227.5
51.39	0.05455	230.2
60.86	0.06520	232.6
Ar(0.800)-NH$_3$(0.200)		
1.24	0.00141	250.3
2.03	0.00231	250.4
5.06	0.00576	251.0
10.13	0.01155	251.7
20.30	0.02342	253.9
36.50	0.04239	256.4
51.44	0.05979	259.4
61.78	0.07247	261.9

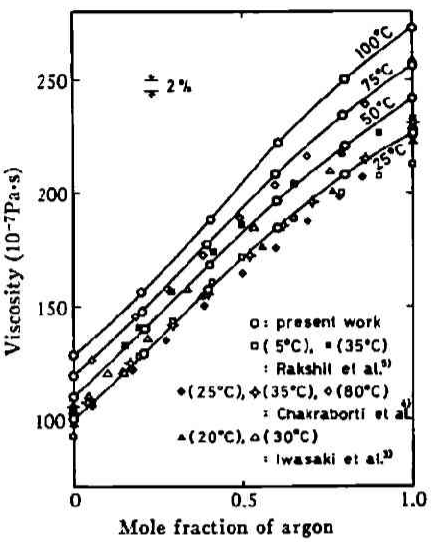

Fig. 3 Viscosity of argon-ammonia system at atmospheric pressure

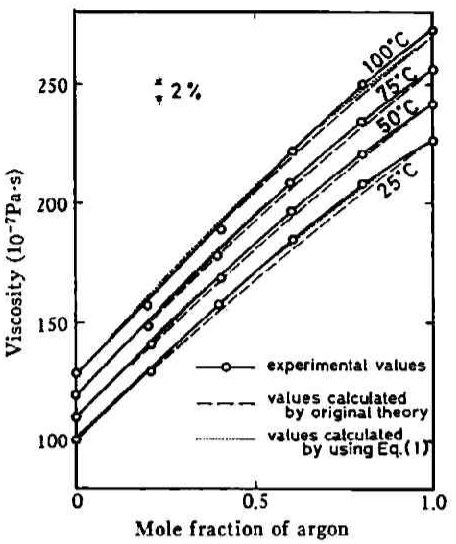

Fig. 4 Comparison of experimental value with original theoretical one and calculated one using Eq. (1) in argon-ammonia system

The viscosity values at atmospheric pressure (1.013×10^5 Pa) were determined in this work through the extrapolation of the viscosity *vs.* pressure curves to 1 atm, and are shown in Fig. 3 in comparison with the other investigations: by Iwasaki *et al.*[3] (20 and 30°C), by Chakraborti *et al.*[4] (25, 35, and 80°C), and by Rakshit *et al.*[5] (5 and 35°C). The values obtained by Iwasaki *et al.*[3] show the similar tendency with those obtained in this work, but those obtained by Chakraborti *et al.*[4] are found to be roughly linear and those obtained by Rakshit *et al.*[5] are seen to be more convex than those obtained in this work.

Discussion

Prediction of gas viscosity for nonpolar-polar mixtures by applying a correction factor

According to Chapman-Enskog's theory[14], the gas viscosity of a dilute binary mixture is given to the first approximation by Eq. (8.2-22) in Ref. 14. If the force constants of pure components are known, the viscosity of binary mixtures can be obtained theoretically. The Lennard-Jones potential function was applied for the nonpolar molecules and the Stockmayer potential function was done for the polar molecules. The force constants of argon were determined by use of the theoretical equation to the third approximation, Eq. (8.2-19) in Ref. 14, and the present experimental viscosity values at

14) J.O. Hirschfelder, C.F. Curtiss and R.B. Bird, "Molecular Theory of Gases and Liquids", John Wiley & Sons, Inc., New York (1954)

atmospheric pressure. The force constants obtained were as follows:

$$\text{Ar:}\quad \sigma_n = 3.44_2[\text{Å}],\quad \varepsilon_n/k = 116[\text{K}]$$

where σ_n and ε_n are the Lennard-Jones potential parameters. The theoretical viscosity of argon $[\eta_1]_1$ required for the calculation of the viscosity of the binary mixtures $[\eta_{mix}]_1$ in Eq. (8.2-22) in Ref. 14 was calculated from Eq. (8.2-19) ($k=3$) using its force constants presented above and that of ammonia $[\eta_2]_1$ had been already calculated in Ref. 2. The force constants between nonpolar and polar molecules were obtained from the combining law, Eqs. (8.6-3) and (8.6-4) in Ref. 14. The theoretical viscosity values of the binary mixtures based on Chapman-Enskog's theory are shown by the dashed line, and the experimental values obtained in this work are denoted by the solid line in Fig. 4. As shown in this figure, the deviation of the theoretical values from the experimental ones is 2% at maximum.

In order to make the deviation smaller, a correction factor was introduced into the combining law as follows[2]:

$$(\varepsilon_{np}/k)' = \varepsilon_{np}/k\ \{(Z_{cn})^{1/4} + (Z_{cp})^{1/4}\}^{-2} \qquad (1)$$

where ε_{np} is a force constant between nonpolar and polar molecular interactions obtained from Eq. (8.6-4) in Ref. 14; k is the Boltzman constant; and Z_{cn} and Z_{cp} are critical compressibility factors of nonpolar and polar substances, respectively. The viscosity values of the gaseous mixtures obtained by applying Eq. (1) are shown by the dotted line in Fig. 4. Apparently the values calculated by using Eq. (1) agree better with the experimental ones than the original theoretical ones do in the argon-ammonia system, as well as in the hydrogen-ammonia system and in the nitrogen-ammonia system[2].

Table 3 Value of constants in Eq. (3)[a]

Temp. (°C)	x_{Ar}	η_0	α'	β'	$\gamma' \cdot 10^{-1}$
25	0.000[b]	101.0	-3.58	-169	1430
	0.208	129.5	-0.379	- 64.2	488
	0.397	157.3	1.71	-218	1050
	0.602	184.7	0.661	31.2	230
	0.800	208.1	0.414	16.1	39.2
	1.000	226.1	0.451	2.39	0.193
50	0.000[b]	110.4	-2.80	- 8.53	268
	0.211	140.3	-0.0321	- 14.1	90.5
	0.403	168.1	1.01	- 54.9	156
	0.600	195.3	0.730	- 8.77	24.3
	0.801	220.6	0.811	- 15.8	29.3
	1.000	241.9	0.394	2.46	0.188
75	0.000[b]	119.8	-2.20	53.6	- 25.0
	0.202	148.0	0.0559	5.33	11.3
	0.394	177.4	0.551	- 4.62	17.8
	0.597	208.1	0.864	- 9.83	14.2
	0.794	234.4	0.537	3.66	4.65
	1.000	256.1	0.494	0.755	0.316
100	0.000[b]	128.9	-1.03	27.3	1.71
	0.200	156.5	-0.0876	18.7	- 5.83
	0.406	188.1	0.556	1.64	0.370
	0.603	221.9	0.893	- 10.8	13.0
	0.800	250.0	0.729	5.12	5.72
	1.000	272.9	0.430	1.72	0.123

a) $\eta = \eta_0(1 + \alpha'\rho + \beta'\rho^2 + \gamma'\rho^3)$ η: 10^{-7} Pa·s ρ: $10^3 \text{kg·m}^{-3} = \text{g·cm}^{-3}$

b) listed in Table 5 in Ref. 1

Initial density dependence of the viscosity for argon-ammonia mixtures

The viscosity of gases under pressures depends on pressure or density, and can be expressed in a power series expansion in density:

$$\eta=\eta_0(1+\alpha'\rho+\cdots) \tag{2}$$

where η_0 is the viscosity extrapolated to zero density and $\alpha'=\eta_0^{-1}(\partial\eta/\partial\rho)_{\rho\to0}$ denotes the initial density dependence of the viscosity[1]. The viscosity of polar gases decreases with increasing pressure or density in lower temperature region, and after having a minimum increases with increasing pressure or density above a certain temperature. For polar gases, the decrease in viscosity is observed in the low pressure or density region below a certain temperature, that is, it is found that α' is negative. In order to investigate the effect of argon concentration upon α' of argon-ammonia mixtures, the gas viscosity was represented by a cubic equation of density as follows:

$$\eta=\eta_0(1+\alpha'\rho+\beta'\rho^2+\gamma'\rho^3) \tag{3}$$

The constants in Eq. (3) were calculated from the experimental viscosity values by means of the least-squares method, and are listed in Table 3. The relation between $\alpha'$and the mole fraction of argon, x_{Ar}, at each temperature is shown in Fig. 5. α' is almost constant above $x_{Ar}=0.6$. Approximating three isothermal points in lower concentration region of argon with a straight line, each

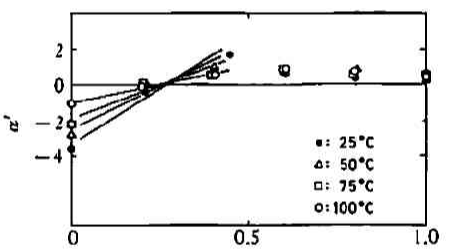

Fig. 5 Initial density dependence of argon-ammonia system

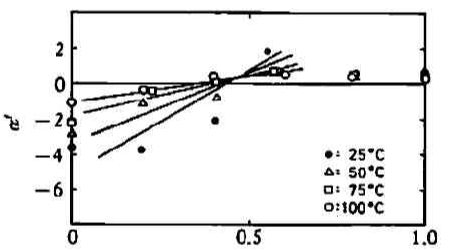

Fig. 6 Initial density dependence of nitrogen-ammonia system

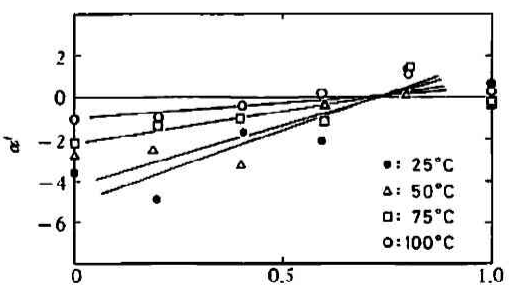

Fig. 7 Initial density dependence of hydrogen-ammonia system

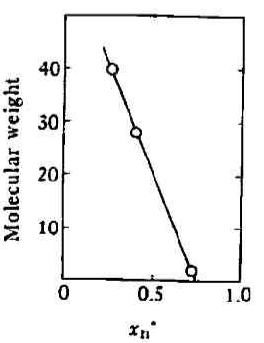

Fig. 8 Relation between molecular weight of nonpolar gas and x_n^*

straight line and $\alpha'=0$ cross at the point $x_{Ar}\fallingdotseq 0.25$, that is, $x_{Ar}^{\circ}=0.25$ where x_{Ar}° is the mole fraction of argon at the point of intersection of straight lines across $\alpha'=0$. The relation similar to Fig. 5 is shown in Figs. 6 and 7 for the nitrogen-ammonia system[1] and for the hydrogen-ammonia system[2], respectively. Figure 6 shows that α' is nearly constant above $x_{N_2}=0.8$, and that $x_{N_2}^{\circ}$ is about 0.4 in the nitrogen-ammonia system. Figure 7 shows that $x_{H_2}^{\circ}$ is about 0.7 in the hydrogen-ammonia system. Thus, it is concluded that α' increases with an increase in temperature and in mole fraction of nonpolar gases, and that the smaller the molecular weight of nonpolar gases is, the larger the x_n° is, where the subscript n denotes a nonpolar gas. Figure 8 shows that an approximately linear relationship exists between the molecular weight of nonpolar geses and the x_n°.

Polar gases show the initial decrease in viscosity below a certain temperature. Singh *et al.*[15] discussed that the anomalous behavior of the viscosity of polar gases was ascribed to molecular association. Therefore, as described in the previous papers[1,2], Figs. 5~7 suggest that a rise in temperature and the presence of nonpolar molecules hinder molecular association of polar gases. Figure 8 shows that the larger the molecular weight of a nonpolar gas is, the smaller the x_n° is. Namely, this figure suggests that the larger the molecular weight of a nonpolar gas is, the more strongly the nonpolar molecules hinder molecular association of polar substances.

The critical temperatures of the gaseous mixtures were determined by taking into account the mole fraction in place of the molecular weight of nonpolar gases, and correlated to α'. The critical temperature of the mixtures was calculated by use of Chueh-Prausnitz's method[16].

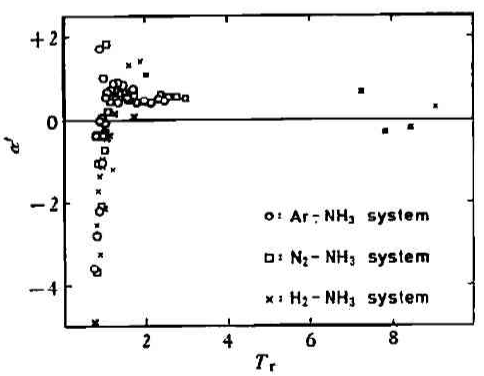

Fig. 9 Correlation between α' and reduced temperature T_r

$$T_{cm}=\sum_j \theta_j T_{cj}+\sum_i\sum_j \theta_i\theta_j\tau_{ij} \quad (4)$$

$$\theta_j=x_j V_{cj}^{2/3}/\sum_j x_i V C_i^{2/3} \quad (5)$$

where T_{cm} is true mixture critical temperature, T_{cj} critical temperature of component j, τ_{ij} an interaction parameter, x_j mole fraction of j, and V_{cj} critical volume of j. Figure 9 shows the

15) Y. Singh, S.K. Deb and A. K. Barua, *J. Chem. Phys.*, **46**, 4036 (1967)

16) R. C. Reid, J. M. Prausnitz and T. K. Sherwood, "The Properties of Gases and Liquids", Third Edition, McGraw-Hill Book Co., New York (1977)

The Review of Physical Chemistry of Japan Vol. 48 No. 2 (1978)

correlation between α' and the reduced temperature, T_r, where $T_r=T/T_{cm}$. In this figure, a similar tendency is observed for the three systems, and this pattern resembles the relations between the reduced second and third virial coefficients and the temperature reduced by the potential parameter[14].

Acknowledgment

The author is sincerely grateful to Professor H. Iwasaki for his continuing interest, encouragement and helpful discussions during this work. The author also wishes to thank Dr. S. Takahashi for many useful suggestions in this paper and Professor T. Makita (Kobe Univ.) for valuable advice with the correlation.

Chemical Research Institute of
Non-Aqueous Solutions
Tohoku University
Sendai
Japan